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Title: RADIOACTIVE TARGETS FOR NEUTRON-INDUCED CROSS SECTION MEASUREMENTS

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RADIOACTIVE TARGETS FOR NEUTRON-INDUCED CROSS SECTION MEASUREMENTS

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Measurements using radioactive targets are important for the determination of key reaction path ways associated with the synthesis of the elements in nuclear astrophysics (s-process), advanced fuel cycle initiative (transmutation of radioactive waste), and stockpile stewardship. High precision capture cross-section measurements are needed to interpret observations, predict elemental or isotopical ratios, and unobserved abundances.

There are two new detector systems that are presently being commissioned at Los Alamos National Laboratory for very precise measurements of (n,γ) and (n,f) cross-sections using small quantities of radioactive samples. DANCE (Detector for Advanced Neutron-Capture Experiments), a 4 π gamma array made up of 160 BaF_2 detectors, is designed to measure neutron capture cross-sections of unstable nuclei in the low-energy range (thermal to ~500 keV) [1]. The high granularity and high detection efficiency of DANCE, combined with the high TOF-neutron flux available at the Lujan Center provides a versatile tool for measuring many important cross section data using radioactive and isotopically enriched targets of about 1 milligram. Another powerful instrument is the Lead-slowing down spectrometer (LSDS), which will enable the measurement of neutron-induced fission cross-section of U-235m and other short-lived actinides in a energy range from 1-200 keV with sample sizes down to 10 nanograms [2]. Due to the short half-life of the U-235m isomer ($T_{1/2}$=26 minutes), the samples must be rapidly and repeatedly extracted from its ^{239}Pu parent. Since ^{239}Pu is itself highly fissile, the separation must not only be rapid, but must also be of very high purity (the Pu must be removed from the U with a decontamination factor >10^{12}). Once extracted and purified, the ^{235m}U isomer would be electrodeposited on solar cells as a fission detector and placed within the LSDS for direct (n,f) cross section measurements.

The production of radioactive targets of a few milligrams will be described as well as the containment for safe handling of these targets at the Lujan Center at LANSCE. To avoid any contamination, the targets are electrochemically fixed onto thin Ti foils and two foils are placed back to back to contain the radioactive material within. This target sandwich is placed in a cylinder made of aluminum with thin translucent windows made of Kapton. Actinides targets, such as ^{234,235,236,238}U, ^{237}Np, and ^{239}Pu are prepared by electrodeposition or molecular plating techniques. Target thicknesses of 1-2 mg/cm^2 with sizes of 1 cm^2 or more have been made. Other targets will be fabricated from separation of irradiated isotopically enriched targets, such as ^{155}Eu from ^{154}Sm, ^{171}Tm from ^{170}Er, and ^{147}Pm from ^{146}Nd, which has been irradiated in the high flux reactor at ILL, Grenoble. A radioactive sample isotope separator (RSIS) is in the process of being commissioned for the preparation of other radioactive targets. A brief summary of these experiments and the radioactive target preparation technique will be given.

[1] M. Heil et al., Nucl. Instr. and Meth. **A459**(2001) 229-246
[2] T. Granier et al., Nucl. Instr. and Meth. **A506**(2003) 149

Radioactive Targets for Neutron-Induced Cross Section Measurement

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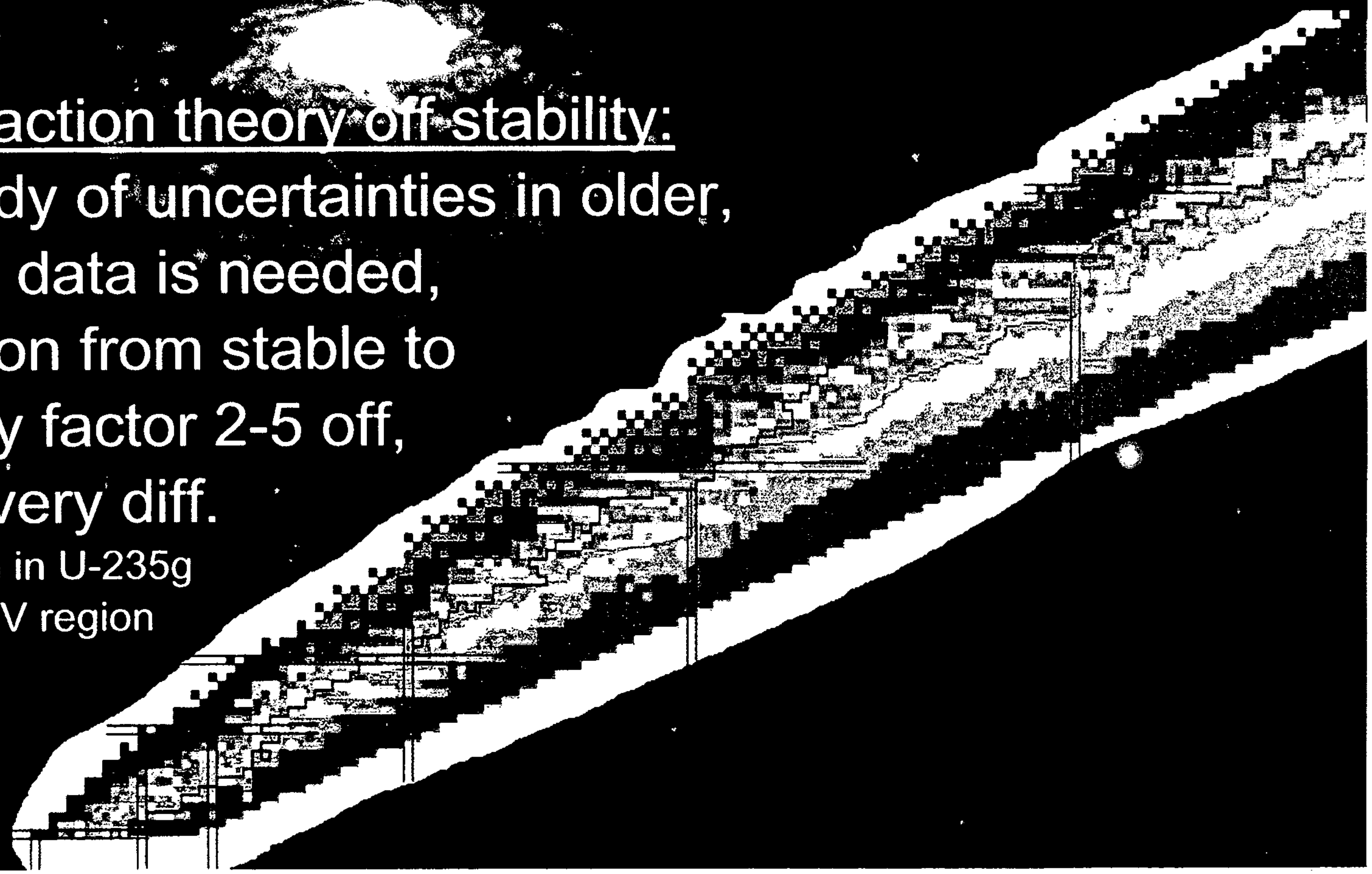
Neutron cross section measurements for:

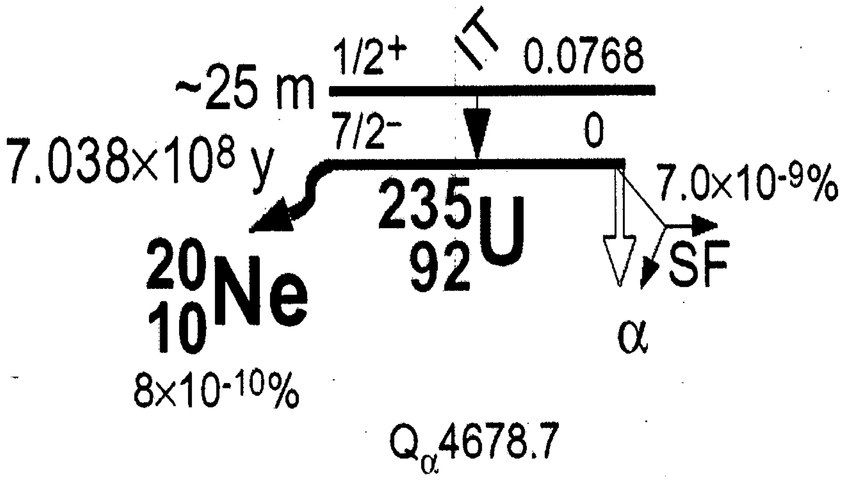
- Nuclear Astrophysics (s-process)
- Reactor physics (IV. Generation)
- Transmutation of nuclear waste (Advanced Fuel Cycle)
- Stockpile Stewardship

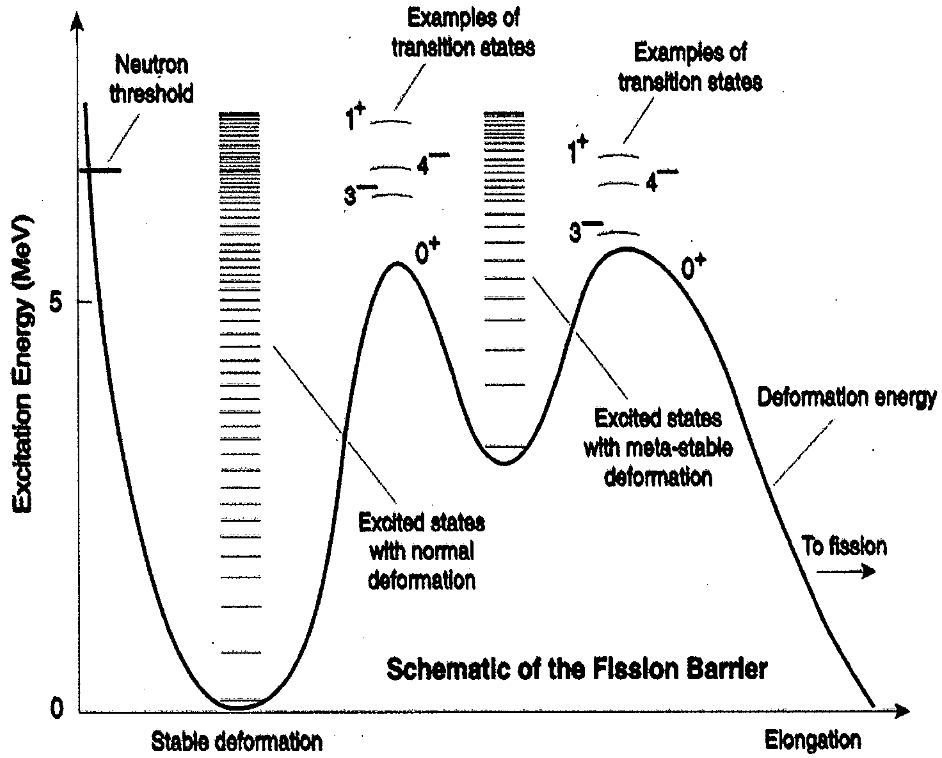
➢ <u>Nuclear reaction theory off stability:</u>
careful study of uncertainties in older, and recent data is needed, extrapolation from stable to unstable by factor 2-5 off, n-capture very diff.

10% in 10keV region in U-235g
239Pu(n,f) 10-14 MeV region



The Isomer U-235m





- 1957 discovery and nuclear characterization
- $T_{1/2}$ = 26.05 ± 0.11 min
- 73 ± 5 eV conversion e-
- 1/2+ state with n --> **+1**; 0
- 7/2- ground state --> –3; –4

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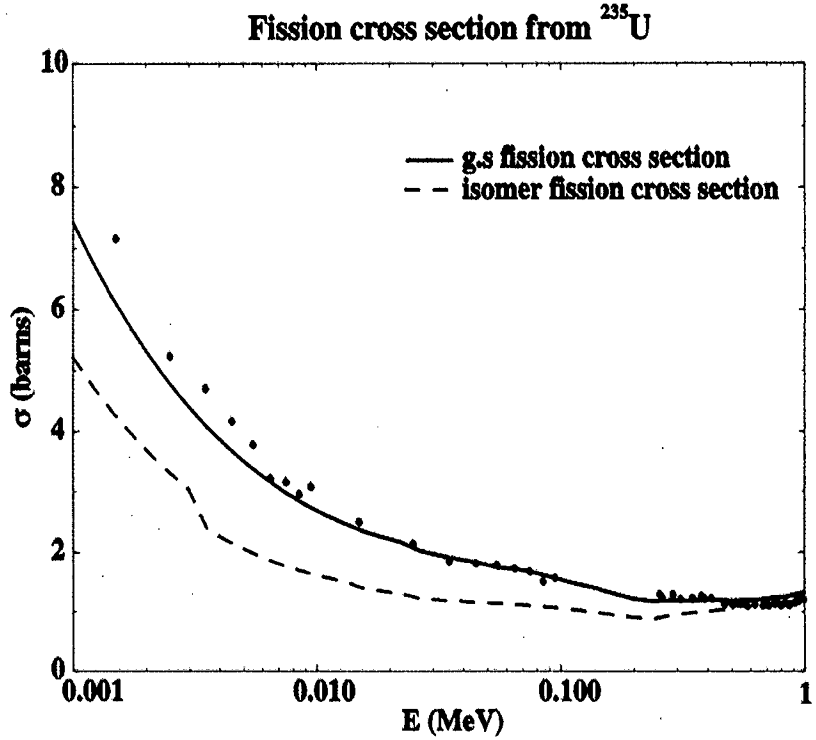
New Measurement

- Calculated cross section at energies relevant to nuclear and stellar explosions lower by up to 30% at energies below 0.5MeV.
- Experiments (LANL, ILL, Dubna) with thermal and cold neutrons
  Talbert W.L. et al., Phys. Rev. C36 (1987) 1896
  $\sigma_m/\sigma_g = 1.42 \pm 0.04$
- Stockpile Stewardship
  ^{235}U (n,n') ^{235m}U on GODIVA
  and ^{235m}U (n,f)



Ref.: J. Eric Lynn, A. C. Hayes, Phys. Rev. C67, 014607 (2003)

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Problem statement: ^{235m}U (n,f)

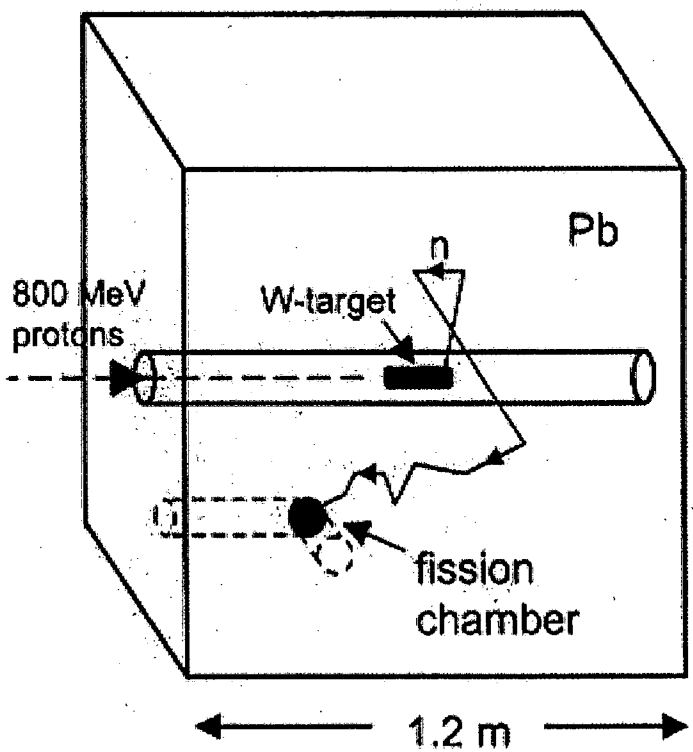
1. ^{239}Pu decay >99% to ^{235m}U
   - separation of ^{235m}U from ^{239}Pu
2. Time scale for the chemistry dominated by $T_{1/2}$ = 26 min
3. Equil. concentration isomer/Pu = 2×10^{-9}
   - about 10 ng of ^{235m}U per 5 g ^{239}Pu
4. Due to the short half-life, this chemistry needs to be done on side at the accelerator facility (but 50 g Plutonium!)
5. Rapidly and repeatedly (every ~90 min), with a very high decontamination factor 10^{12}
6. Target substrate must be thin to enable detection of fission fragments.

Lead slowing down spectrometer

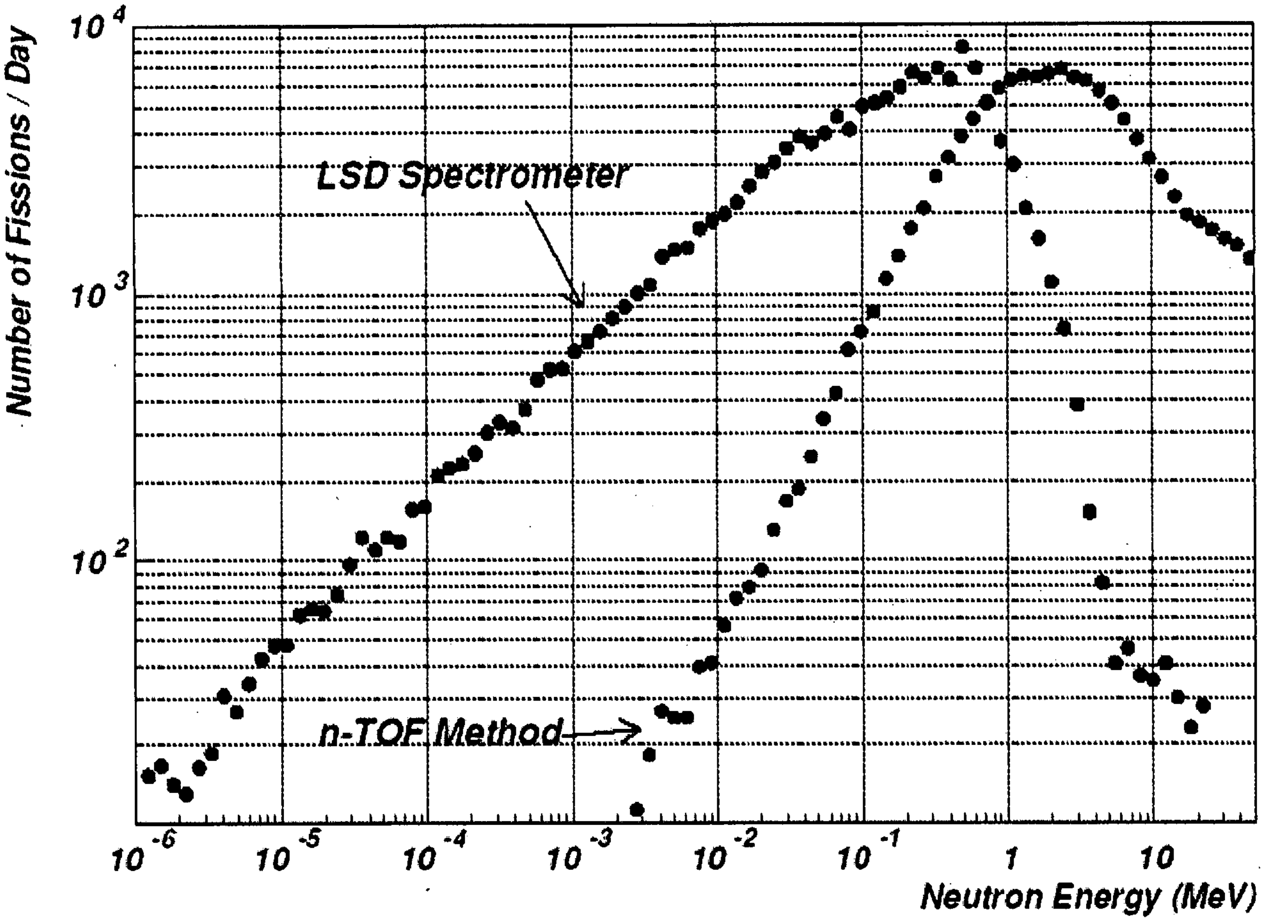
- ng samples need high neutron flux
- LSDS works by „recycling“ neutrons (stores like in a bottle)
- 20 t lead cube increases neutron flux
  by ~1000 (ΔE_n~30%)
- neutron spallation source is pulsed
- For E_n < 100 keV

$$\langle E_n \rangle = K / (t + t_0)^2$$

- This is proven technology at electron Linacs, van de Graaff accelerators. Spallation sources should be much better.



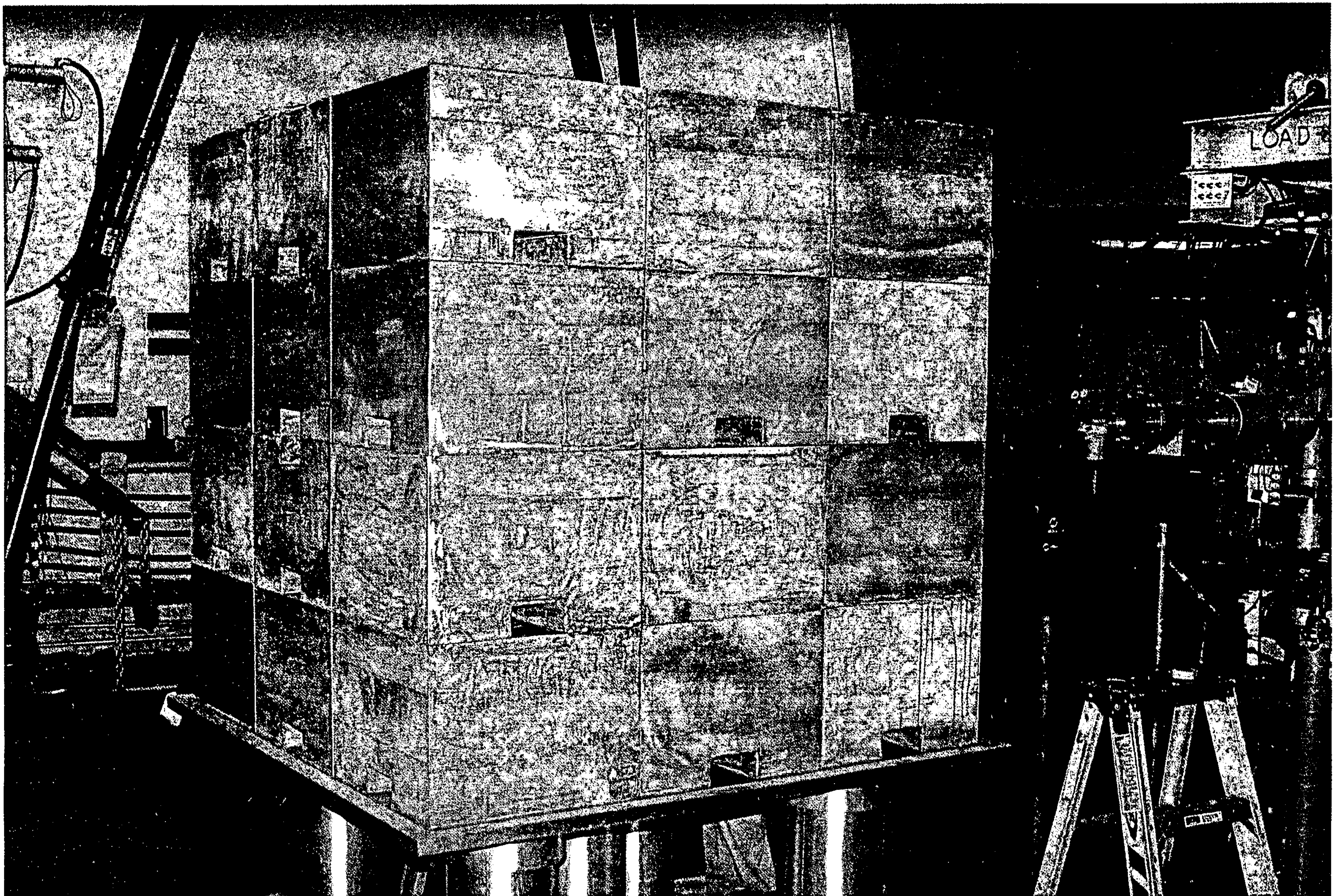
Estimated ^{235m}U Fission Yield



- 10 ng ^{235m}U target
- 1 μA 800 MeV beam + LSDS
- 100 μA beam + n-TOF (100x too optimistic)

L. Pangault *et al.*, LA-UR-02-7718;

T. Granier *et al.*, NIM A<u>506</u>(2003) 149



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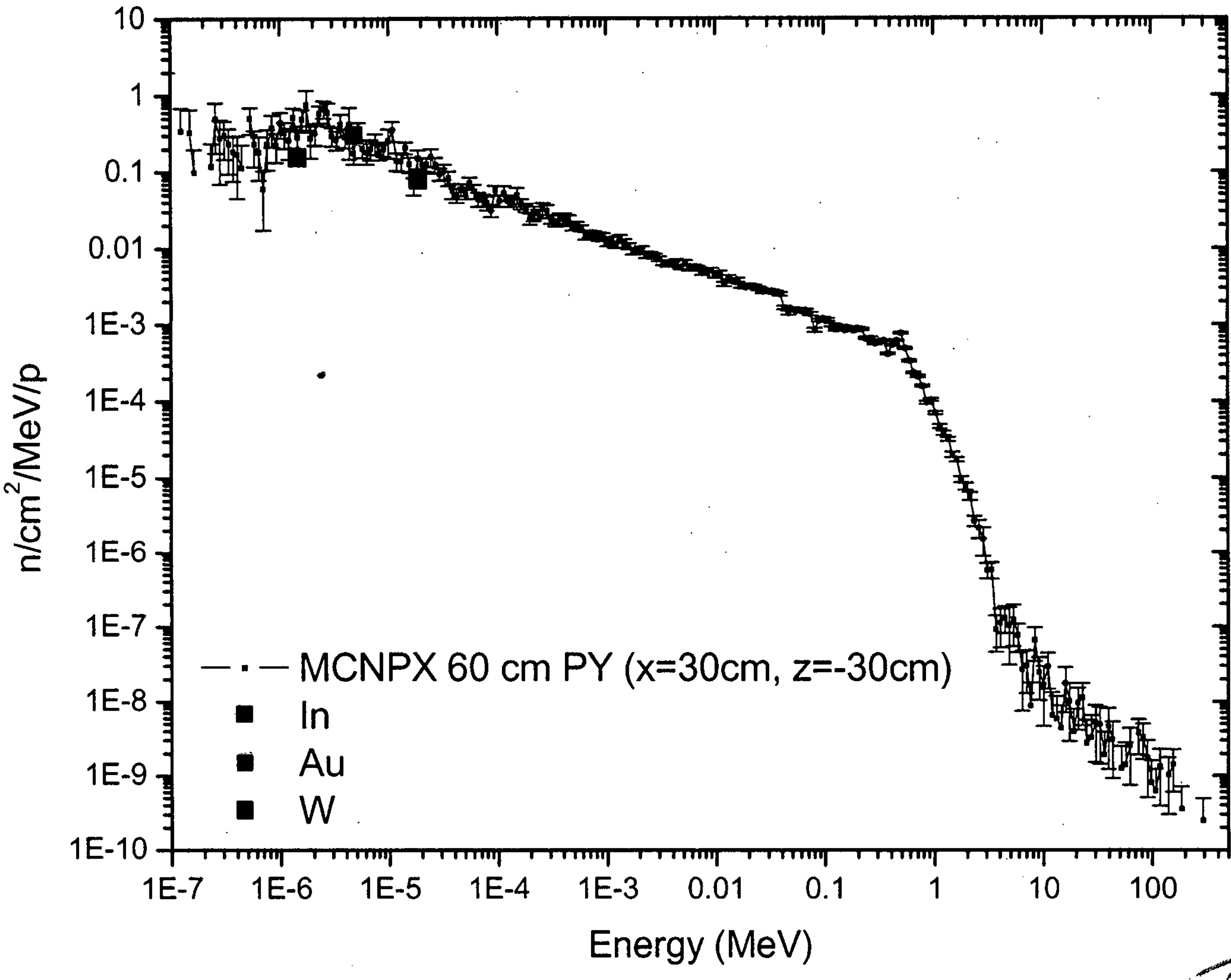
Neutron spectrum of the LSDS (W spallation target)



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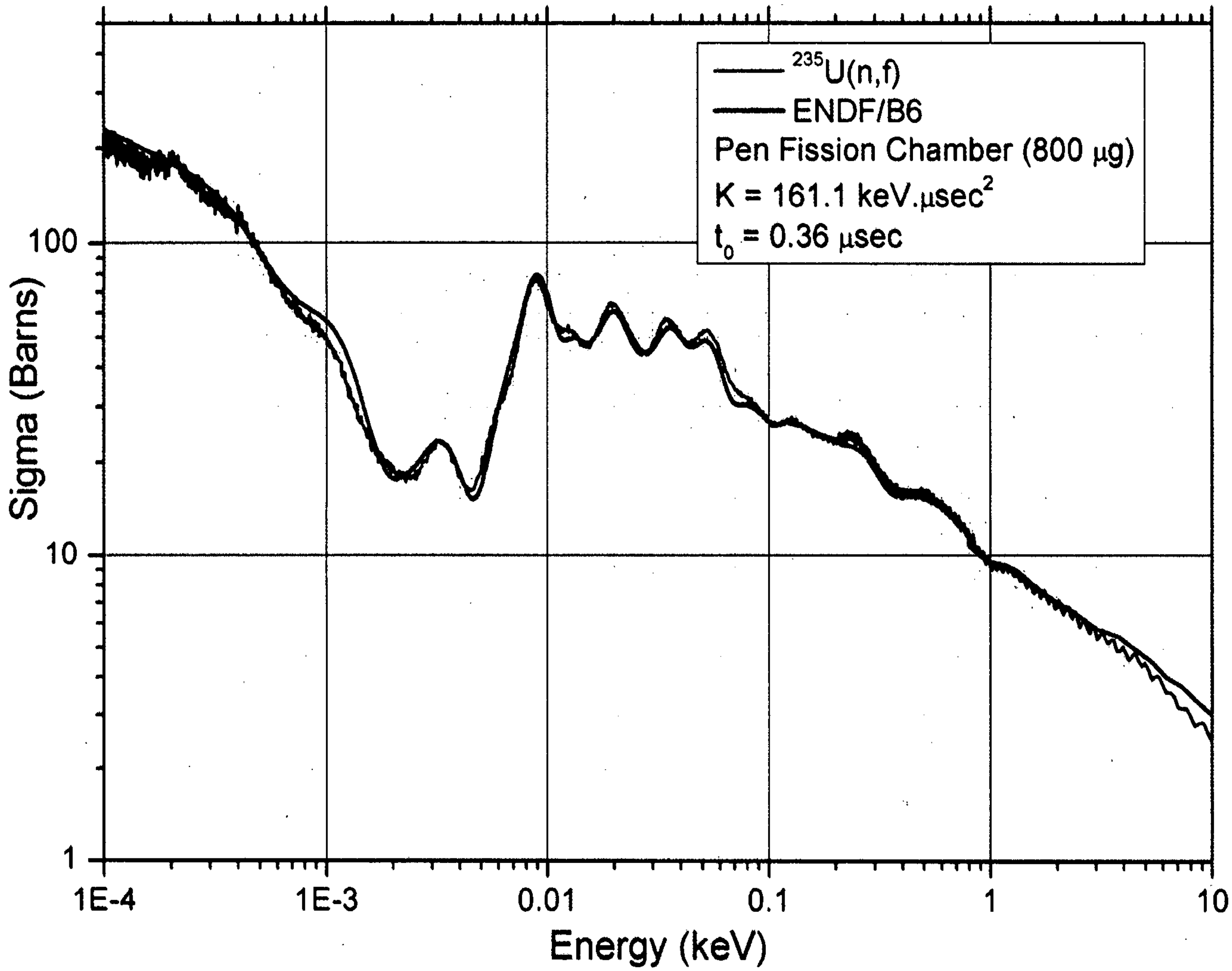
Preliminary results with lower beam current – detector tests



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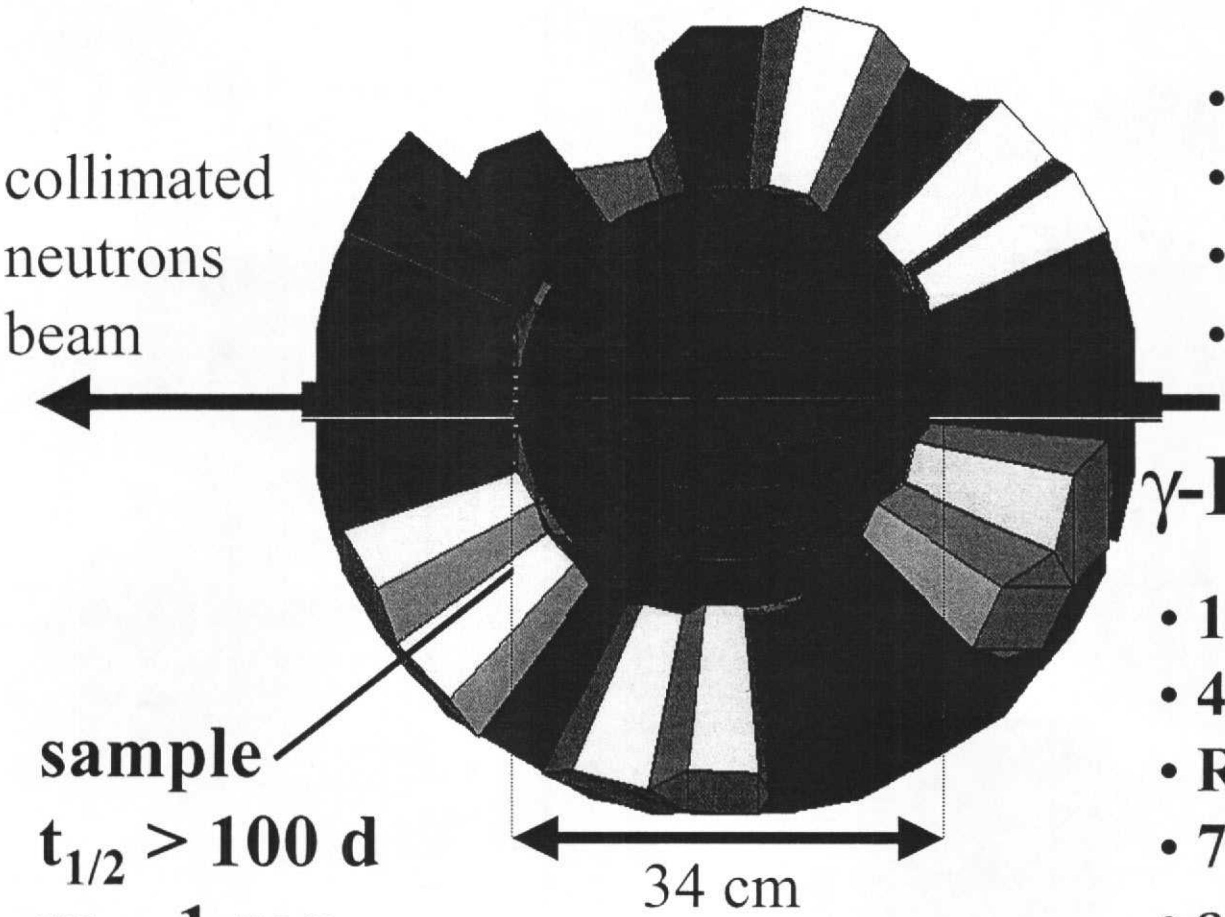
*D*etector for *A*dvanced *N*eutron *C*apture *E*xperiments



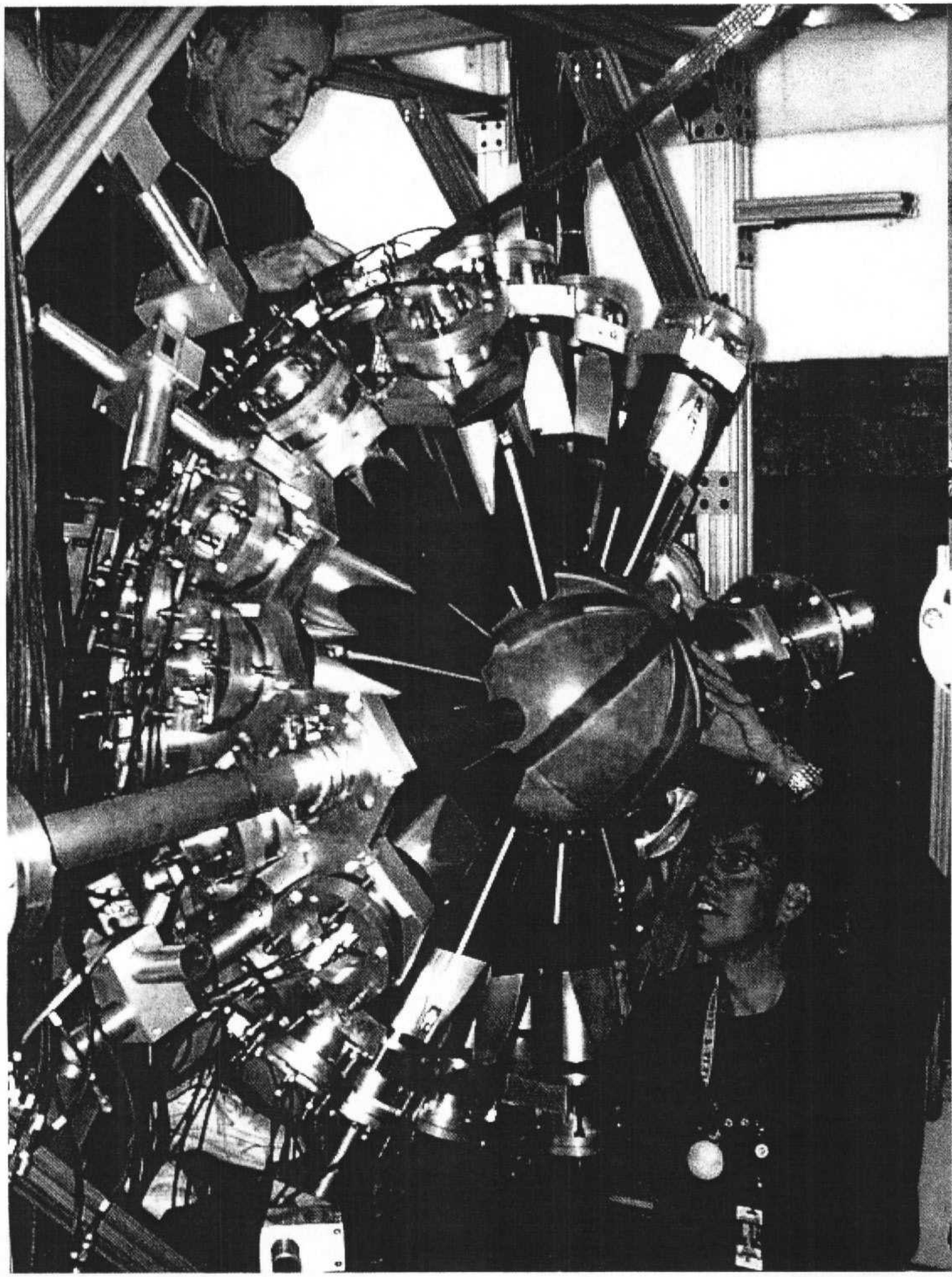
$t_{1/2}$ > 100 d
m ~ 1 mg

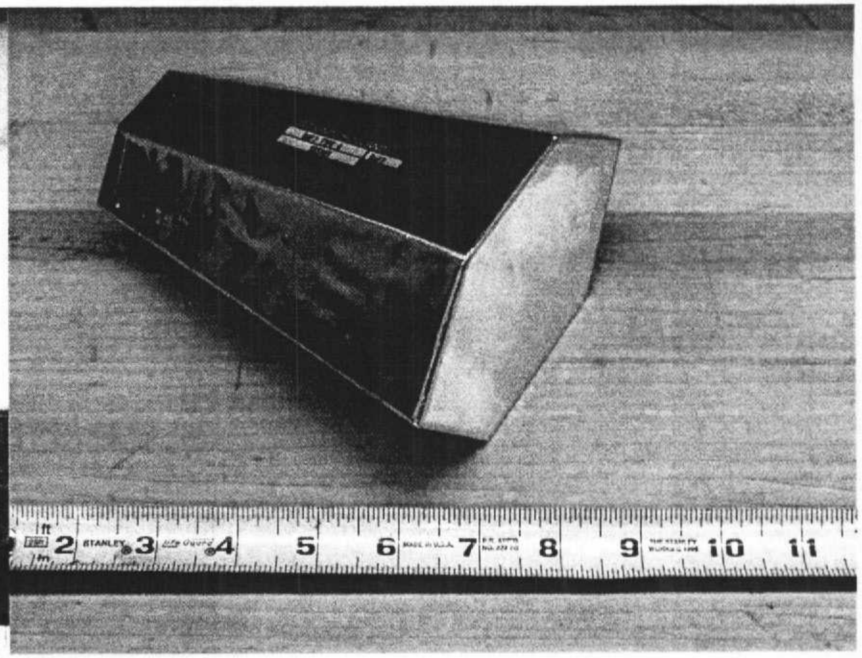
neutrons:

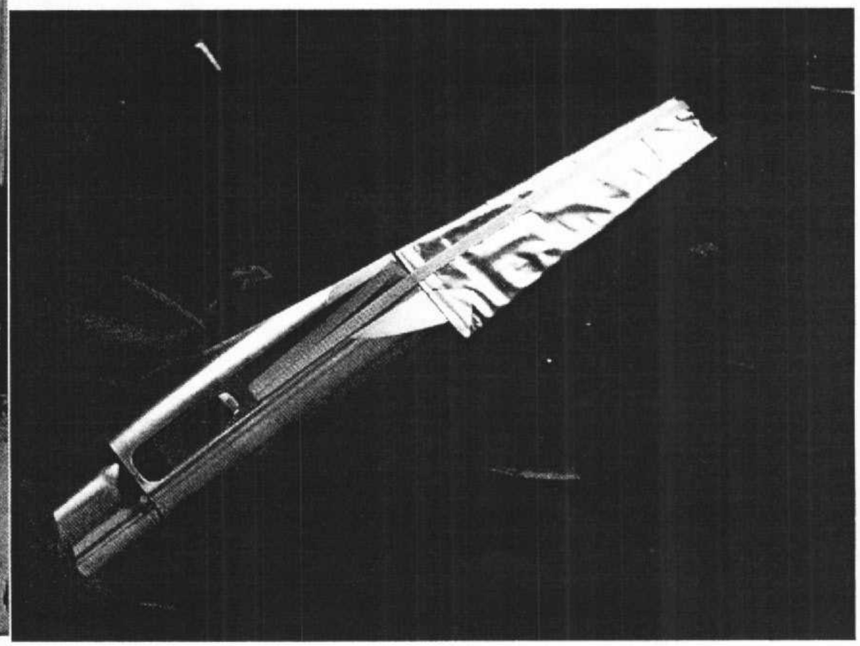
- **spallation source**
- **thermal .. 500 keV**
- **20 m flight path**
- **$3*10^5$ n/s/cm^2/decade**

γ-Detector:

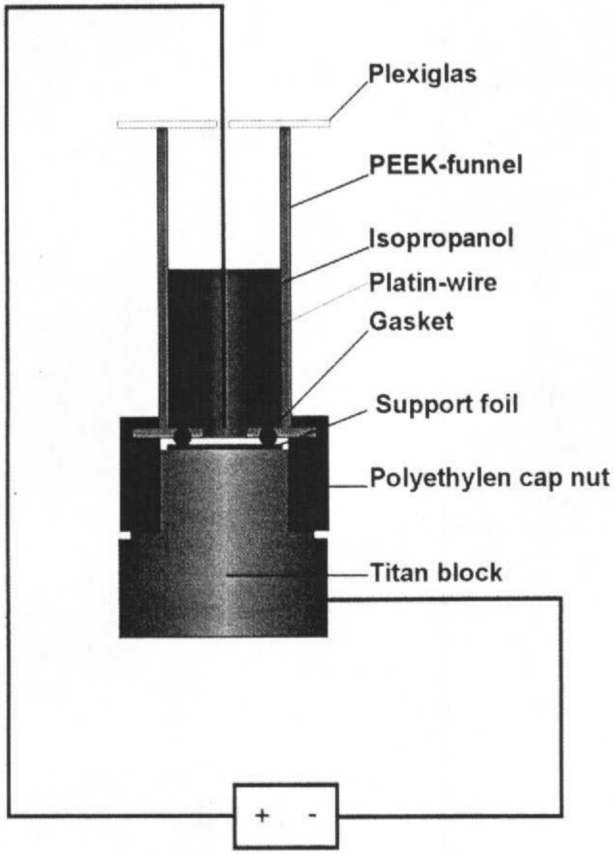
- **160 BaF_2 crystals**
- **4 different shapes**
- **R_i=17 cm, R_a=32 cm**
- **7 cm ^6LiH inside**
- **$\varepsilon_\gamma \approx$ 90 %**
- **$\varepsilon_{casc} \approx$ 98 %**




STANLEY



Target preparation for Uranium



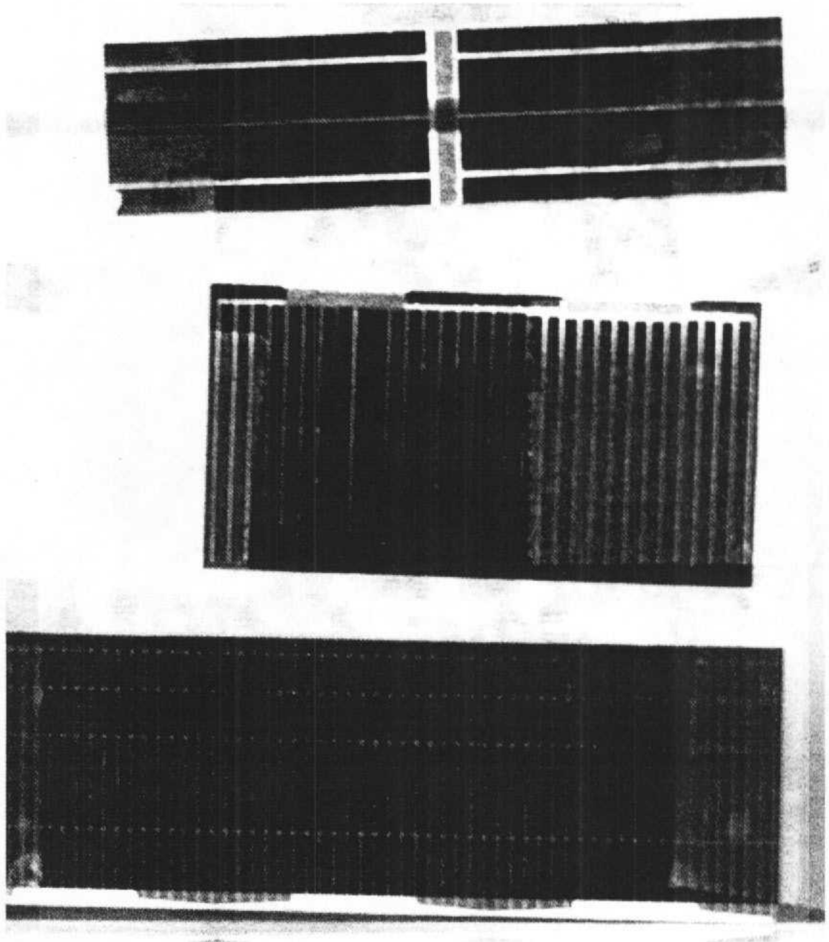
- Uranylnitrate in 0.1 M HNO_3
- 5 μL with 14 mL isopropanol
- Strong cooling required
- 10 min with 100 V, then further with 200, 300, 400, 600, and 800V (makes 60 min)
- 2 cm^2 targets with 50-60 % yield
- 700 mg/cm^2 or 2 mg/cm^2 in two runs

- Approx. 2.5 mL of isopropanol
- 100-150 μL of 0.1 M HNO_3
- 100-150 μL of 0.1 M HCl
- 30 mA for 30 minutes
  (U is 120 to 60 V)

= for plating on solar cells

solar cells as fission detector

- ✓ Energy threshold 30 MeV
- ✓ Puls linearity
- ✓ 65-90% detection efficiency
- ✓ 2-20% energy resolution
- ✓ Mass resolution 8 amu
- ✓ Position resolution 5 to 9 mm
- ✓ Time resolution 10 ns (FWHM)
- \+ Price
- \+ Cut to any shape
- \+ No vacuum needed
- \+ No n dose effect observed
- \+ Low sensitivity to α, β, γ, n
- \+ No HV bias required
- \- Fragility
- \- Noise / light sensitivity
- \- Lifetime vs. α activity



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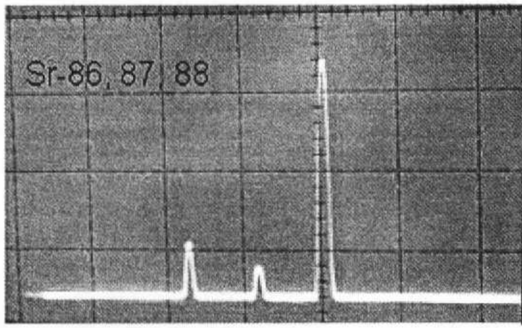
Reactor-produced samples

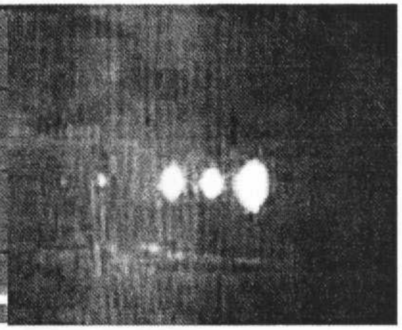
isotopically enriched targets have been irradiated @ ILL Grenoble, F
The samples have been irradiated for 41 days at a neutron flux of $1.5 * 10^{15}$ n/cm^2 s. After a cooling period of 4 months the dose rate and produced amounts are given in the following table:

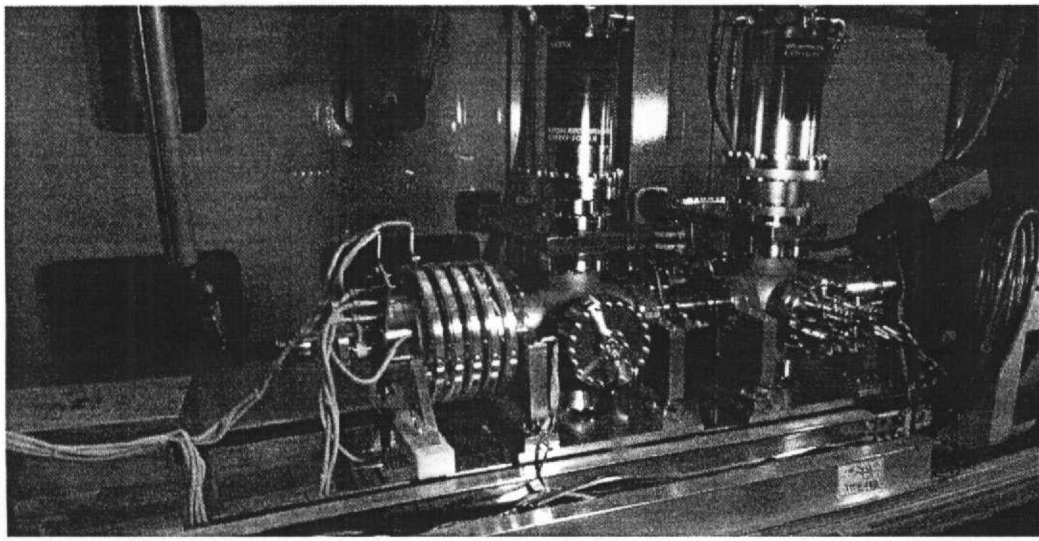
Target	Amount irradiated	Cross section /barn	product	Amount produced	Dosis (after cooling)	Half-life
Nd-146	221 mg	1.4	Pm-147	3.1 mg	0.03 Sv/h (2.9 Ci)	2.62 a
Sm-154	196 mg	7.2	Eu-155	7.3 mg	1 Sv/h (3.5 Ci)	4.76 a
Er-170	250 mg	6.0	Tm-171	7.7 mg	0.7 Sv/h (8.5 Ci)	1.92 a

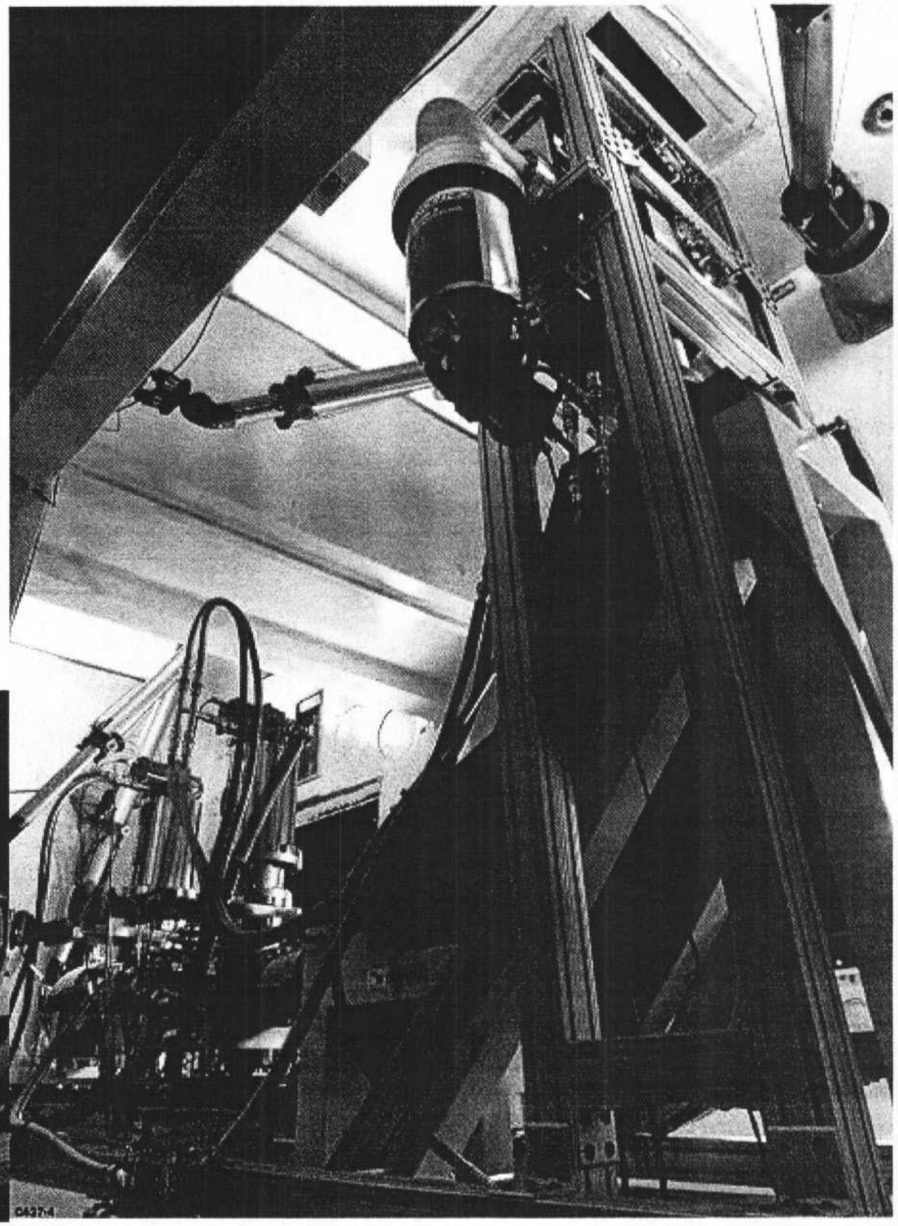
Radioactive Species Isotope Separator (RSIS)

- Has been assembled and tested in a double hot cell at the CMR building at LANL **(~50 μA separator in a hot cell)**
- Samples has to be radiochemically separated from Al implated foils

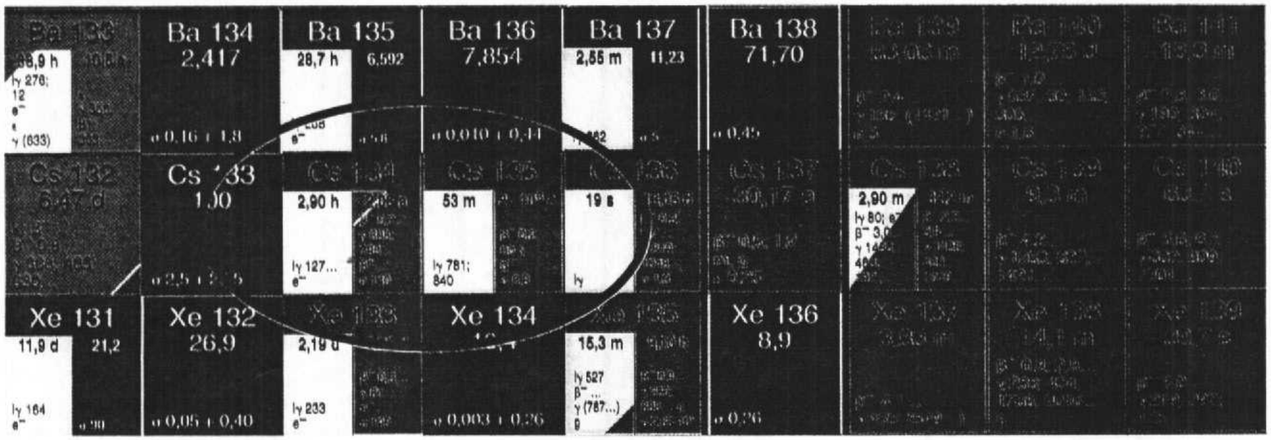


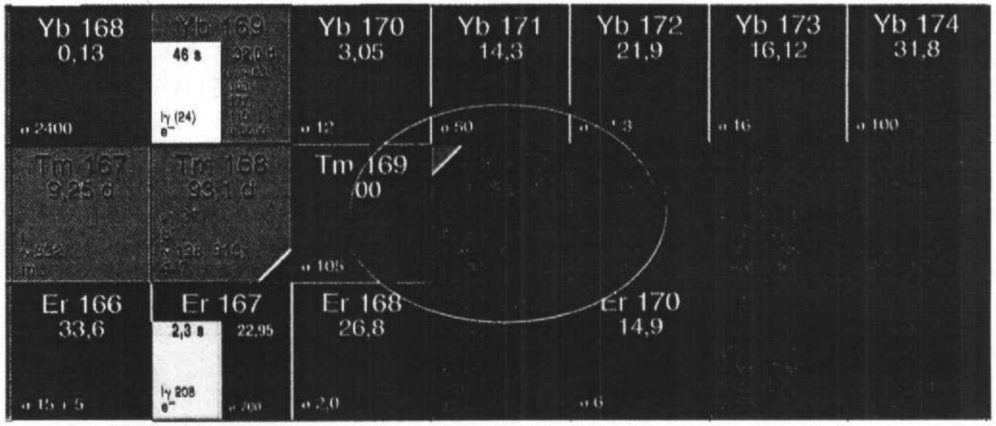






Mass-separated radioactive samples





Near future plans

- $^{239,240,242}Pu$
- ^{241}Am
- ^{171}Tm
- ^{155}Eu
- ^{147}Pm
- ^{135}Cs

and many other radioisotopes

Isotopes for Future Study define a multi-year program

s-branch point	w/o DANCE	With DANCE
Ni-63	-	√
Se-79	-	√
Kr-81,85	-	√
Cs-134,135	-, ~	√
Nd-147	-	-
Pm-147,148	~, -	√, -
Sm-151	√	√
Gd-153	-	√
Eu-154,155	-, ~	√
Tb-160	-	√
Ho-163	-	√
Tm-170, 171	-, √	√
Ta-179	-	√
W-185	-	√
Tl-204	-	√

Other targets of interest:

$^{93,95}Zr$, ^{99}Tc, ^{152}Eu, ^{176}Lu, ^{186}Re, ^{193}Pt, ^{205}Pb, ^{86}Rb, ^{88}Y, ^{94}Nb, ^{107}Pd, ^{141}Ce, (^{119}Sn, ^{102}Pd) ....

Stockpile Stewardship Targets:

$^{170,171}Tm$, $^{232,234,235,236,238}U$, ^{155}Eu,^{238}Pu, ^{241}Am, $^{173,174,176}Lu$, ^{192}Ir

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